

‘C₆₀ spin-charging’ with an eye on a quantum computer

J P Connerade[§] and V K Dolmatov[¶]

[§] The Blackett Laboratory, Imperial College, Prince Consort Road, London SW7 2BZ, UK E-mail: Jean-Patrick@Connerade.com

[¶] Department of Physics and Earth Science, University of North Alabama, Florence, AL 35632, USA. E-mail: vkdolmatov@una.edu

Abstract. A question whether there exists an interaction between the spins of the endohedral atom $A@C_{60}$ and the properties of the confining shell which might affect the alignment of, or manipulation by, the spins for building a register for a quantum computer is discussed. It is argued that an effect, termed the ‘C₆₀ spin-charging’ effect, can occur in endohedral atoms and would affect the operation of a quantum register. The effect is exemplified by choosing the 3d (Cr and Mn) and 4d (Mo and Tc) transition metal atoms as well as a rare-earth Eu atom as the case study. A class of high-spin atoms which are less suitable for building a quantum register is, thus, identified.

PACS numbers: 03.67.Lx, 31.90.+s, 81.07.Nb, 85.35.Be

Submitted to: *J. Phys. B: At. Mol. Opt. Phys.*

The use of a non-zero spin atom confined by C₆₀ (referred to as the endohedral A@C₆₀ atom) as the building block of the register for a quantum computer was proposed by Harneit [1]. Obviously, the higher the spin of the atom, the better. Confined atoms then must be atoms with one or more multielectron semifilled subshells in their configuration whose electron spins are aligned. The study by Harneit [1] focused on the use of a semifilled shell N(2p³)@C₆₀ atom. The general idea for building the register for a quantum computer depends on the freedom to align the spin of the encapsulated atom, on the ability of the C₆₀ confining cage to screen the spins from the influence of unwanted decohering fields and on the ability to write (read) to (from) an assembly of confined atoms held together as an array.

It is, therefore, interesting to explore whether, in fact, the freedom to align the spins of encapsulated atoms exists independently of the properties of a confining shell and whether external fields are able to perturb this alignment. The latter question has already been addressed theoretically by Connerade and Solov'yov [2] and Amusia and Baltenkov [3] who studied the properties of a spherical C₆₀ cage and showed under what conditions the C₆₀ screening of an external field remains effective. The former of the two questions is addressed in the present paper by accounting for an effect termed the ‘C₆₀ spin-charging effect’.

The C₆₀ spin-charging effect was recently uncovered as a by-product by Dolmatov *et al* [4] in the study of e⁻ + A@C₆₀ electron elastic scattering. The quintessence of the effect is that both the electron spin-up $P_{n\ell\uparrow}(r)$ and spin-down $P_{n\ell\downarrow}(r)$ functions of a high spin encapsulated atom *A*, such as an atom with one or more multielectron semifilled subshells in its configuration, may be drawn noticeably, but very differently, into the region of the C₆₀ wall. This results in loading the C₆₀ cage with electron density of a preferred spin orientation. Naturally, the effect is accompanied by the loss of some electron spin-density localized on the confined atom *A* itself. Clearly, the phenomenon is potentially important for the proposed realization of an A@C₆₀ register for a quantum computer. It is the ultimate aim of the present paper to delineate the spin-charging effect more precisely for this purpose. To meet this goal, the 3d-, 4d- and 4f-semifilled shell Cr(...3d⁵4s¹, ⁷S), Mn(...3d⁵4s², ⁶S), Mo(...4d⁵5s¹, ⁷S), Tc(...4d⁵5s², ⁶S) and Eu(...4f⁷6s², ⁸S) atoms encapsulated inside C₆₀ are chosen for the completeness of the case study.

In the following, we briefly outline the methodology to calculate the C₆₀ spin-charging effect in an endohedral semifilled shell atom, A@C₆₀.

A convenient way to account for the structure of a semifilled shell atom is provided by the spin-polarized Hartree-Fock approximation (SPHF) developed by Slater [6]. SPHF accounts for the fact that spins of all electrons in a semifilled subshell of the atom (e.g., in the 3d⁵ subshell of Mn) are co-directed, in accordance with Hund’s rule, say, all pointing upward. This results in splitting of a closed $n\ell^{2(2\ell+1)}$ subshell in the atom into two semifilled subshells of opposite spin orientations, $n\ell^{2\ell+1}\uparrow$ and $n\ell^{2\ell+1}\downarrow$. This is in view of the presence of exchange interaction between $n\ell\uparrow$ electrons with only spin-up electrons in the original semifilled subshell of the atom (like the 3d⁵ \uparrow subshell in the Mn atom) but absence of such for $n\ell\downarrow$

electrons. Thus, the SPHF configurations of Cr, Mn, Mo, Tc and Eu are as follows: Cr(...3p³↑3p³↓3d⁵↑4s¹↑, ⁷S), Mn(...3p³↑3p³↓3d⁵↑4s¹↓, ⁶S), Mo(...4p³↑4p³↓4d⁵↑5s¹↑, ⁷S), Tc(...4p³↑4p³↓4d⁵↑5s¹↓, ⁶S) and Eu(...4d⁵↑4d⁵↓4f⁷↑...6s¹↑6s¹↓, ⁸S). SPHF equations for the ground state of a semifilled shell atom differ from ordinary HF equations for closed shell atoms by accounting for exchange interaction only between electrons with the same spin orientation (↑, ↑ or ↓, ↓) [6, 7]. To model a A@C₆₀ atom, we account for the presence of the C₆₀ cage by adding a rectangular (in the radial coordinate r) potential well $U_{C_{60}}(r)$ of a finite width Δ , depth U_0 and inner radius r_0 to the HF (SPHF) equations [5], as in many of other studies, see, e.g., [4, 8, 9, 10, 11] and references therein:

$$U_{C_{60}}(r) = \begin{cases} -U_0, & \text{if } r_0 \leq r \leq r_0 + \Delta \\ 0 & \text{otherwise.} \end{cases} \quad (1)$$

In the literature, some inconsistency is present in choosing the magnitudes of Δ , U_0 and r_0 of the C₆₀ phenomenological potential (1), cp., e.g., References [5, 4, 8, 9, 10, 11] with each other. In the present paper, following [10], we choose $\Delta = 2.9102$ au (which is twice of the covalent radius of carbon), $r_0 = 5.262$ au = $R_c - 1/2\Delta$ ($R_c = 6.7173$ au being the radius of the C₆₀ skeleton) and $U_0 = 7.0725$ eV (which was found by matching the electron affinity $EA = -2.65$ eV of C₆₀ with the assumption that the orbital momentum of the 2.65-eV-state is $\ell = 1$ [10]). This choice is most consistent with observations. Calculated $P_{ns\uparrow}(r)$ and $P_{ns\downarrow}(r)$ functions of valence spin-up and spin-down electrons of the Cr, Mn, Mo, Tc and Eu atoms, both free and encapsulated inside C₆₀, are depicted in figure 1.

Note how the encapsulation of the chosen atoms inside the C₆₀ cage draws their outer $P_{ns\uparrow}(r)$ and, respectively, $P_{ns\downarrow}(r)$ orbital functions into the region of the C₆₀ wall. This implies a significant transfer of electron density from the encapsulated atom to the cage, but, more importantly in the context of the present paper, a transfer of electron *spin-density* from the atom to the cage. The transfer makes the cage become ‘spin-charged’. The C₆₀ cage becomes spin-charged even for the *spin-neutral* 4s² and 5s² subshells of endohedral Mn and Tc, respectively. This is because of the stronger drain of the valence $ns\downarrow$ than $ns\uparrow$ electron density from the atom to the cage. Interestingly enough, the spin-dependent drain of the valence electron density does not emerge in Eu@C₆₀ where both the $P_{6s\uparrow}(r)$ and $P_{6s\downarrow}(r)$ orbital functions are drawn into the C₆₀ cage equally strongly, in contrast to the outer $P_{ns\uparrow}(r)$ and $P_{ns\downarrow}(r)$ orbital functions of the endohedral Mn and Tc atoms. This is because the semifilled 4f⁷↑ subshell of Eu lies much deeper relative to its 6s¹↑ and 6s¹↓ subshells than the semifilled $nd^{5\uparrow}$ subshell of Mn ($n = 3$) or Tc ($n = 4$) relative their valence $(n + 1)s^{1\uparrow}$ and $(n + 1)s^{1\downarrow}$ subshells. For this reason, the exchange interaction between the 4f↑ and 6s↑ electrons in Eu is negligibly small. Hence, there is practically no difference between the $P_{6s\uparrow}(r)$ and $P_{6s\downarrow}(r)$ functions both in free and encapsulated Eu. As a result, the Eu atom retains its electron *spin-density* intact upon confinement inside C₆₀, which could prove important for an eventual application.

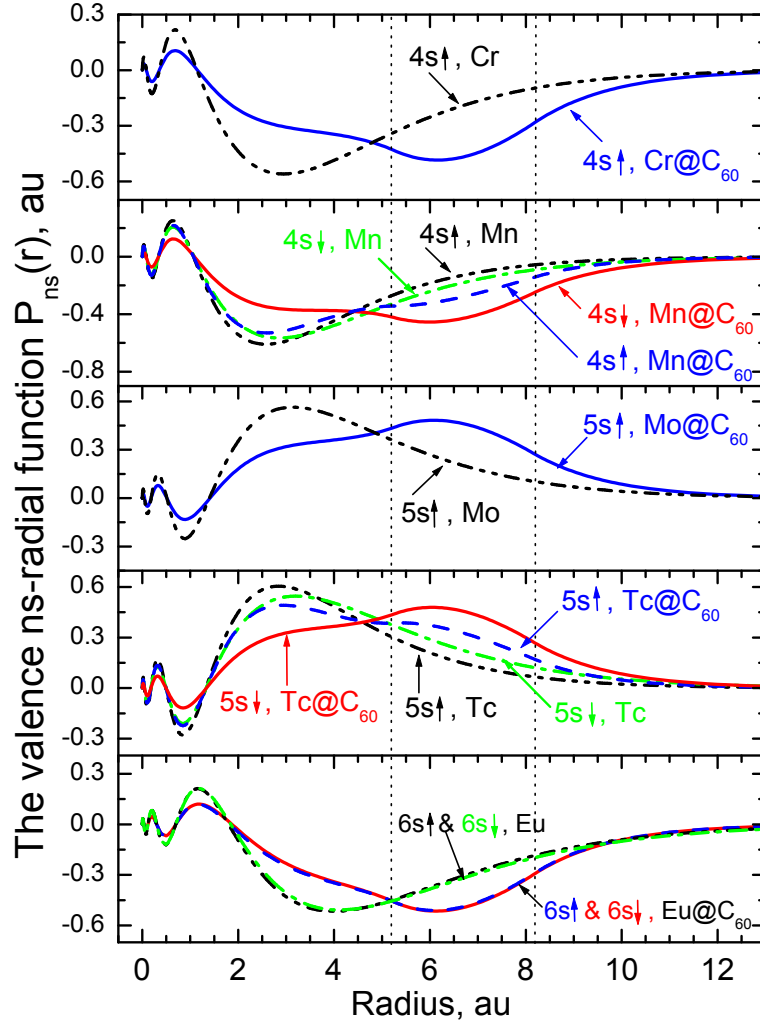


Figure 1. (Color online) Calculated $P_{ns\uparrow}(r)$ and $P_{ns\downarrow}(r)$ radial functions (in atomic units, au) of the valence subshells of the Cr@C₆₀, Mn@C₆₀, Mo@C₆₀, Tc@C₆₀ and Eu@C₆₀ atoms versus those of the free atoms, as marked. Vertical dotted lines locate the position of the C₆₀ wall, $5.262 \leq r \leq 8.17$ au.

In conclusion, the authors believe that the C₆₀ spin-charging effect we have described will affect the manipulation of spins in the corresponding A@C₆₀ systems and that it must inhibit, or at least render more complex, the operation of a quantum register. The present paper thus brings to light a class of high-spin atoms which are less suitable for building a quantum register, namely those which are subject to a strong electron spin-density drain from the atom to the C₆₀ cage.

Acknowledgments

VKD acknowledges the supported of NSF Grant no. PHY-1305085. The undergraduate students C. J. Bayens, M. B. Cooper and M. E. Hunter of the University of North Alabama are thanked for their assistance with the calculations.

References

- [1] Harneit W 2002 Fullerene-based electron-spin quantum computer *Phys. Rev. A* **65** 032322
- [2] Connerade J P and Solov'yov V A 2005 Dynamical screening of a confined atom by a fullerene *J. Phys. B: At. Mol. Opt. Phys.* **38** 807
- [3] Amusia M Ya and Baltenkov A S 2006 Effect of plasma oscillations of C₆₀ collectivized electrons on photoionization of endohedral noble-gas atoms *Phys. Rev. A* **73** 062723
- [4] Dolmatov V K, Cooper M B and Hunter M E 2014 Electron elastic scattering off endohedral fullerenes A@C₆₀: the initial insight *J. Phys. B: At. Mol. Opt. Phys.* **47** 115002
- [5] Connerade J P, Dolmatov V K and Manson S T 1999 A unique situation for an endohedral metallofullerene *J. Phys. B: At. Mol. Opt. Phys.* **32** L395
- [6] Slater J C 1974 *The Self-Consistent Field for Molecules and Solids* (New York: McGraw-Hill)
- [7] M. Ya. Amusia and L. V. Chernysheva, 1997 *Comput. Atomic Processes: A Handbook for the ATOM Programs* (Bristol: Institute of Physics Publishing)
- [8] Dolmatov V K and Keating D A 2012 Xe 4d photoionization in Xe@C₆₀, Xe@C₂₄₀ and Xe@C₆₀@C₂₄₀ *J. Phys.: Conf. Ser.* **208** 022097
- [9] Pushka M J and Nieminen R M 1993 Photoabsorption of atoms inside C₆₀ *Phys. Rev. A* **47** 1181
- [10] Winstead C and McKoy V 2006 Elastic electron scattering by fullerene, C₆₀ *Phys. Rev. A* **73** 012711
- [11] Phaneuf R A, Kilcoyne A L D, Aryal N B, Baral K K, Esteves-Macaluso D A, Thomas C M, Hellhund J, Lomsadze R, Gorczyca T W, Ballance C P, Manson S T, Hasoglu M F, Schippers S and Müller 2013 A Probing confinement resonances by photoionizing Xe inside a C₆₀⁺ molecular cage *Phys. Rev. A* 053402